

Evaluation and modelling of transient bulk dynamics during the supercritical CO₂ drying of aerogels

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Production of aerogels starting from almost all precursors requires a supercritical drying step to preserve the mesoporous structure of the gel. Such systems include most of the organic gels derived from synthetic polymers and almost all biopolymers. To understand, optimize, scale-up and control the supercritical CO₂ drying process, several mathematical models have in the past been developed and fitted to experimental data. Since there is a lack of correlations describing the convective-dispersive transport of the solvent during the supercritical drying process, the existing models assume an ideal behavior of the bulk domain. Thus, they are likely to be overfitted and may not represent the real drying dynamics but rather biased and thus of limited practical applicability.

Nowadays, most of the measurement systems track the concentration of solvent at the outlet of the autoclave. The obtained signal is a convolution of the mass transport occurring in the gel and the convection and dispersion of the solvent from the surface of the gel to the detector. The convective-dispersive transport of the solvent through the system dominates the signal, hiding the true nature of the drying dynamics. Decoupling the main transport processes has been proven to be a difficult task due to the transient kinetic nature of the process. In order to decouple the different mass transport processes occurring during the process and to generate data that is equipment independent, new experimental and modeling techniques are required to successfully describe the complex dynamics of supercritical CO₂ drying.

A dynamic kernel deconvolution technique based on the Aris-Taylor equation was developed to recover the bulk dynamics of the system in the form of mean residence time and dispersion coefficient. Two correlations were found and globally fitted to a large number of experiments performed on a wide range of process conditions relevant to the industrial supercritical CO₂ drying. It was found that gravity plays a critical role on the process dynamics giving rise to two distinctive bulk dynamic regimes during the supercritical CO₂ drying: fluid stratification and dispersion due to density gradients. It is also demonstrated that the residence time of the process and the dispersion coefficient varies considerably during the drying. Residence time values in the range of 500 s to 2000 s and dispersion coefficients 40 times larger than the molecular diffusion coefficient of the solvent in the supercritical CO₂ clearly shows the impact that bulk dynamics has on the overall kinetics of the supercritical CO₂ drying process.

The obtained correlations are used to validate and adjust a finite element model describing the bulk dynamics using the diffusion-advection equation coupled to the Navier-Stokes equations. The effect of gravity on the system is implemented as an artificial diffusion coefficient streamlined to the velocity field and which magnitude is a function of the density gradient parallel to the gravity vector. This implementation increased the convergence rate of the model, but most importantly reduced the computation time from days to minutes compared to implementing the gravity effect as a volume force term in the Navier-Stokes equations.